

# Enhancing Mixing and Diffusion with Plastic Flow

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We use numerical simulations to examine two-dimensional particle mixtures that strongly phase separate in equilibrium. When the system is externally driven in the presence of quenched disorder, plastic flow occurs in the form of meandering and strongly mixing channels. In some cases this can produce a fast and complete mixing of previously segregated particle species, as well as an enhancement of transverse diffusion even in the absence of thermal fluctuations. We map the mixing phase diagram as a function of external driving and quenched disorder parameters.

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There have been a growing number of experiments on collections of small particles such as colloids moving over periodic or complex energy landscapes generated by various optical methods [1, 2, 3, 4, 5, 6, 7, 8] or structured surfaces [9]. Such static and dynamical substrates can produce a variety of new particle segregation mechanisms [2, 4, 6, 7] as well as novel types of logic devices [3]. Driven particles on periodic substrates can also exhibit enhanced diffusive properties such as the recently proposed giant enhancement of the diffusion which occurs at the threshold between pinned and sliding states [7, 10, 11, 12, 13, 14]. This enhancement has been demonstrated experimentally for colloids moving over a periodic optical substrate [7] and could be important for applications which require mixing and dispersing of different species of particles [7]. A limiting factor for using diffusion enhancement to mix particles is that the diffusion is enhanced only in the direction of the external drive. For instance, in a two-dimensional system with a corrugated potential that is tilted in the direction of the corrugation barriers, there is no enhancement of the diffusion in the direction transverse to the corrugation barriers at the pinned to sliding threshold. It would be very valuable to identify a substrate that allows for strong enhancement of the diffusion in the direction transverse to the tilt of the substrate, or one that would facilitate the mixing of particle species that are intrinsically phase separated in equilibrium. Such a substrate could be used to perform fast mixing of species and would have applications in microfluidics, chemical synthesis, and creation of emulsions and dispersions.

In this work we show that a phase separated binary assembly of interacting particles in the presence of a two-dimensional random substrate tilted by a driving field undergoes rapid mixing and has an enhancement of the diffusion transverse to the tilt direction. The motion of the particles occurs via *plastic flow* in the form of meandering channels which have significant excursions in the direction perpendicular to the drive, leading to mixing of the two particle species. The mixing and diffusion occur

even in the absence of thermal fluctuations and arise due to the complex multi-particle interactions. We map the mixing phase diagram as a function of external drive and substrate properties and identify regimes of rapid mixing. We find that as the difference between the two particle species increases, the mixing becomes increasingly asymmetric with one species penetrating more rapidly into the other. Our work shows that plastic flow can be used as a mechanism for mixing applications, and also provides a new system for the study of collective dynamical effects.

We simulate a two-dimensional system with periodic boundary conditions in the  $x$  and  $y$  directions containing two species of Yukawa particles labeled  $A$  and  $B$  with charges  $q_A$  and  $q_B$ , respectively. The particle-particle interaction potential between particles  $i$  and  $j$  of charges  $q_i$  and  $q_j$  at positions  $\mathbf{r}_i$  and  $\mathbf{r}_j$  is  $V(r_{ij}) = E_0 q_i q_j \exp(-\kappa r_{ij})/r_{ij}$ , where  $E_0 = Z^{*2}/4\pi\epsilon\epsilon_0$ ,  $\epsilon$  is the dielectric constant,  $Z^*$  is the unit of charge,  $\kappa$  is the screening length, and  $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ . We fix  $\kappa = 4/a_0$  where  $a_0$  is the unit of length in the simulation. The system size is  $L = 48a_0$ . The motion of particle  $i$  is determined by integration of the overdamped equation of motion

$$\eta \frac{d\mathbf{r}_i}{dt} = \mathbf{F}_i^{cc} + \mathbf{F}_i^s + \mathbf{F}_d \quad (1)$$

where  $\eta$  is the damping term which is set equal to unity. Here  $\mathbf{F}_i^{cc} = -\sum_{i \neq j}^N \nabla V(r_{ij})$  is the particle-particle interaction force, where  $N$  is the total number of particles in the system. The particle density is  $\rho = N/L^2$ . The substrate force  $\mathbf{F}_i^s = -\sum_{k=1}^{N_p} \nabla V_p(r_{ik})$  comes from  $N_p$  parabolic trapping sites placed randomly throughout the sample. Here  $V_p(r_{ik}) = -(F_p/2r_p)(r_{ik} - r_p)^2 \Theta(r_p - r_{ik})$ , where  $F_p$  is the pinning strength,  $r_p = 0.2a_0$  is the pin radius,  $r_{ik} = |\mathbf{r}_i - \mathbf{r}_k^{(p)}|$  is the distance between particle  $i$  and a pin at position  $\mathbf{r}_k^{(p)}$ , and  $\Theta$  is the Heaviside step function. The pin density is  $\rho_p = N_p/L^2$ . The external driving force  $\mathbf{F}_d = F_d \hat{\mathbf{x}}$  is applied uniformly to all the particles. The units of force and time are  $F_0 = E_0/a_0$  and  $\tau = \eta/E_0$ , respectively. We neglect thermal fluc-

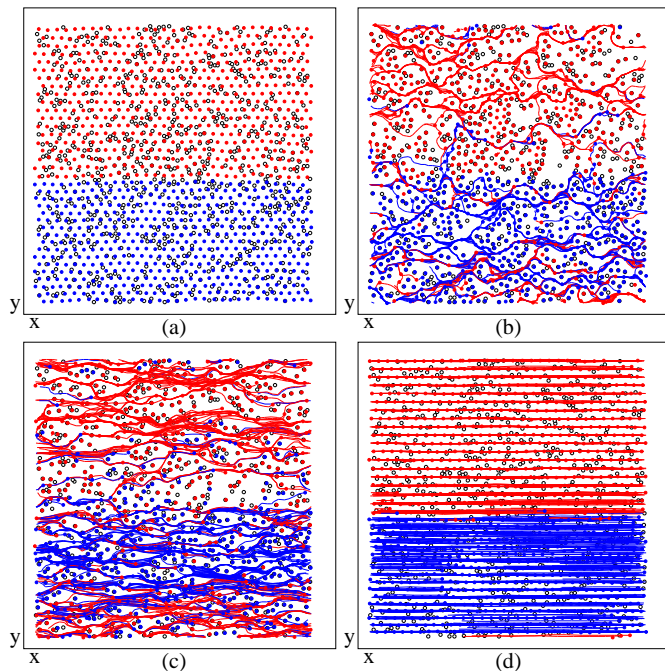


FIG. 1: Red circles and red lines: particle positions and trajectories for species *A*; blue circles and blue lines: particle positions and trajectories for species *B*; open black circles: pinning site locations in a system with particle density  $\rho = 0.7$ , pin density  $\rho_p = 0.34$ , and pinning force  $F_p = 1.0$  at different driving forces. (a)  $F_d = 0.0$ , (b)  $F_d = 0.1$ , (c)  $F_d = 0.4$ , and (d)  $F_d = 1.1$ .

tuations so that  $T = 0$ . If the two particle species are initialized in a phase separated state, in the absence of an external drive and disorder the particles will not mix unless the temperature is raised above melting.

In Fig. 1(a) we show the initial phase separated particle configuration for a 50:50 mixture of the two particle species with  $q_A/q_B = 3/2$  and  $q_A = 3$ . The particles are placed in a triangular lattice of density  $\rho = 0.7$  which is immediately distorted by the pinning sites of density  $\rho_p = 0.34$  and strength  $F_p = 1.0$ . Species *A* occupies a larger fraction of the sample due to its larger charge  $q_A$  and correspondingly larger lattice constant compared to species *B*. An external driving force  $F_d$  is applied in the  $x$ -direction and held at a fixed value.

Figure 1(b) illustrates the particle trajectories at  $F_d = 0.1$  over a period of  $10^5$  simulation steps. The trajectories form meandering riverlike structures with significant displacements in the direction transverse to the drive, producing intersecting channels that permit species *A* to mix with species *B*. When the trajectories and particle positions are followed for a longer period of time, the amount of mixing in the system increases. The riverlike channel structures are typical of plastic flow of particles in random disorder, where a portion of the particles are temporarily trapped at pinning sites while other particles move past, so that the particles do not

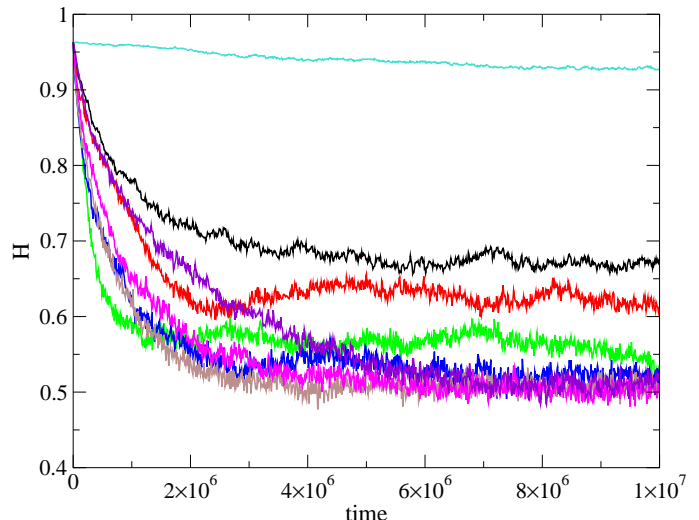


FIG. 2: Measure of local homogeneity  $H$  vs time for the system in Fig. 1 at  $F_d = 0.05$  (black),  $0.1$  (red),  $0.25$  (green),  $0.4$  (blue),  $0.5$  (brown),  $0.6$  (magenta),  $0.7$  (violet), and  $1.1$  (top curve).  $H = 1$  for phase segregation and  $H = 0.5$  for complete mixing.

keep their same neighbors over time. This type of plastic flow has been observed in numerous one-component systems including vortices in type-II superconductors [15, 16, 17, 18, 19, 20, 21], electron flow in metal dot arrays [22], and general fluid flow through random disorder [23, 24]. These works have shown that by changing the strength and size of the disorder, the amount of transverse wandering or tortuosity of the riverlike channels can be adjusted, and that these channels appear even for  $T = 0$  [16, 17, 18, 19]. In our system we measure the diffusion in the  $y$ -direction,  $d_y = |\langle \mathbf{r}_i(t) \cdot \hat{\mathbf{y}} - \mathbf{r}_i(0) \cdot \hat{\mathbf{y}} \rangle|^2$ , and find a long time transverse diffusive motion with  $d_y(t) \propto t^\alpha$  and  $\alpha = 1.0$ , indicative of normal diffusion. Single component systems exhibiting plastic flow also show a similar transverse diffusive behavior [17]. The diffusion in our system is not induced by thermal motion but rather occurs due to the complex many-body particle interactions that give rise to the meandering riverlike channels. In Fig. 1(c) we plot the particle trajectories in the same system at  $F_d = 0.4$ . At this drive, a larger fraction of the particles are mobile and the riverlike channels become broader. As the drive is further increased, all the particles are depinned, the meandering riverlike structures are lost, and the mixing of the particles decreases. Such a state is shown in Fig. 1(d) at  $F_d = 1.1$ . For higher values of  $F_d > 1.1$ , flow similar to that shown in Fig. 1(d) appears.

In order to quantify the mixing, for each particle we identify the closest neighboring particles by performing a Voronoi tessellation on the positions of all particles in the system. We then determine the probability  $H$  that a particle is of the same species as its neighbors. If the sys-

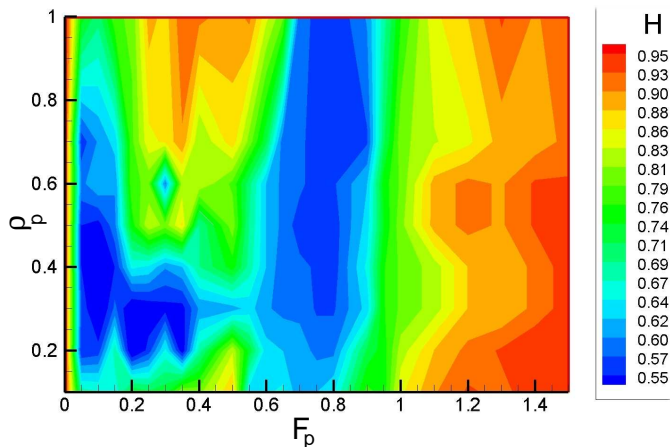


FIG. 3: Mixing phase diagram of pinning density  $\rho_p$  vs pinning strength  $F_d$  in the form of a height map of the local homogeneity  $H$  obtained from a series of simulations with  $F_p = 1.0$  and particle density  $\rho = 0.7$ . Strong mixing regions are blue and weak mixing regions are red.

tem is thoroughly mixed, the local homogeneity  $H = 0.5$ , while if it is completely phase separated,  $H$  is slightly less than one due to the boundary between the two species. In Fig. 2 we plot  $H(t)$  for the system in Fig. 1 at different values of  $F_d$  ranging from  $F_d = 0.05$  to  $F_d = 1.1$ . For the lower drives  $F_d \leq 0.1$ , there are few channels and a portion of the particles remain pinned throughout the duration of the simulation so that mixing saturates near  $H = 0.6$  to  $0.7$ . For the intermediate drives  $0.1 < F_d \leq 0.5$  any given particle is only intermittently pinned, so at long times all the particles take part in the motion and the system fully mixes, as indicated by the saturation of  $H$  to  $H = 0.5$ . For drives  $0.5 < F_d < 0.9$  the system can still completely mix but the time to reach full mixing increases with  $F_d$ . At  $F_d > 0.9$  where the particles are completely depinned, the mixing becomes very slow as shown by the  $H(t)$  behavior for  $F_d = 1.1$ . Within the strongly mixing regime,  $H(t) \propto A \exp(-t)$  at early times before complete mixing occurs.

In Fig. 3 we plot the mixing phase diagram of pinning density  $\rho_p$  versus driving force  $F_d$  as determined by the local homogeneity  $H$  obtained from a series of simulations with  $F_p = 1.0$  and  $\rho = 0.7$ . The value of  $H$  is measured after  $3 \times 10^7$  simulation time steps. Blue indicates strong mixing and red indicates weak mixing. For  $F_d > 1.0$  and all values of  $\rho_p$ , all of the particles are moving in a fashion similar to that illustrated in Fig. 1(d). Since the plastic flow is lost, mixing is very inefficient in this regime. For  $F_d < 0.6$  at high pinning densities  $\rho_p > 0.7$ , most of the particles are pinned, preventing a significant amount of mixing from occurring. A region of strong mixing appears at  $0.6 < F_d < 0.9$  for all values of  $\rho_p$ . Here, the particles intermittently pin and depin, producing the large amount of plastic motion necessary to generate mixing. There is another strong region of mix-

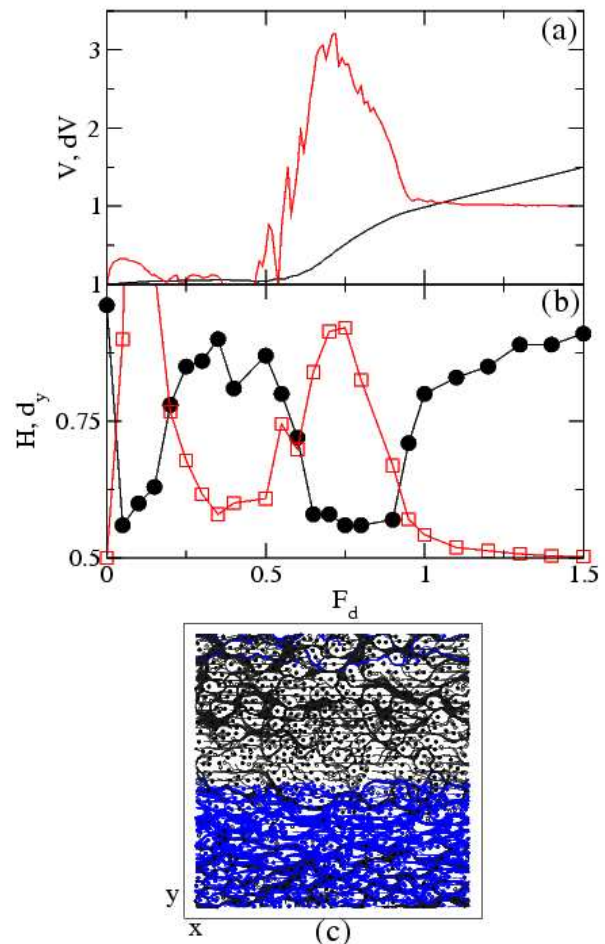


FIG. 4: (a) Black line: the average particle velocity  $V$  vs  $F_d$  for a system with  $f_p = 1.0$ ,  $\rho_p = 0.34$ , and  $\rho = 0.7$ . Red line: the corresponding  $dV/dF_d$  curve. (b) Black circles: local homogeneity  $H$ ; red squares: net transverse displacement  $d_y$  for the same system as in (a). The high mixing regime ( $H < 0.6$ ) is correlated with enhanced transverse displacements and the peak in  $dV/dF_d$ .  $d_y$  has been shifted down for presentation purposes. (c) Particle positions (circles) and trajectories for species A (black) and species B (blue) in a system with  $q_A/q_B = 3$  and  $F_d = 0.2$ . The mixing is asymmetric with species A moving into the region occupied by species B before species B moves into the area occupied by species A.

ing for lower pinning densities  $0.2 < \rho_p < 0.4$  and low  $F_d < 0.4$ . In this regime there are more particles than pinning sites so that interstitial particles, which are not trapped by pinning sites but which experience a caging force from neighboring pinned particles, are present. At low drives the interstitial particles easily escape from the caging potential and move through the system; however, the pinned particles remain trapped so that the interstitial particles form meandering paths through the pinned particles. This result shows that even a moderately small amount of disorder combined with a small drive can generate mixing. As the pinning density is further decreased

to  $\rho_p < 0.15$ , the amount of mixing also decreases.

In Fig. 4(a) we demonstrate how the mixing phases are connected to the transport properties of the system by plotting the net particle velocity  $V = \langle N^{-1} \sum_{i=1}^N \mathbf{v}_i \cdot \hat{\mathbf{x}} \rangle$  and  $dV/dF_d$  versus driving force  $F_d$  for a system with  $\rho_p = 0.34$  and  $F_p = 1.0$ . Here  $\mathbf{v}_i$  is the velocity of particle  $i$ . In Brownian systems, it was previously shown that an enhanced diffusion peak is correlated with a peak in the derivative of the velocity force curve [10, 11, 12, 13, 14]. Figure 4(a) shows that there is a peak in  $dV/dF_d$  spanning  $0.5 < F_d < 0.9$  which also corresponds to the region of high mixing in Fig. 3. There is also a smaller peak in  $dV/dF_d$  at small drives  $F_d < 0.2$  produced by the easy flow of interstitial particles. For  $F_d > 1.0$ ,  $V$  increases linearly with  $F_d$  since the entire system is sliding freely. In Fig. 4(b) we plot the local homogeneity  $H$  for the same system taken from the phase diagram in Fig. 3. The maximum mixing ( $H < 0.6$ ) falls in the same region of  $F_d$  where the peak in  $dV/dF_d$  occurs. Figure 4(b) also shows that the net traverse particle displacement  $d_y$  has peaks in the strong mixing regimes.

We have also examined the effect of significantly increasing  $q_A/q_B$  so that the system is even more strongly phase separated. In general, we find the same mixing features described previously; however, the time required for complete mixing to occur increases with increasing  $q_A/q_B$ . The mixing also becomes *asymmetric*: the more highly charged species  $A$  invades the region occupied by species  $B$  before the less highly charged species  $B$  spreads evenly throughout the sample. In Fig. 4(c) we illustrate the particle trajectories during the first  $3 \times 10^6$  simulation time steps for a system with  $q_A/q_B = 3$  at  $F_d = 0.2$ . The mixing asymmetry can be seen from the fact that the black trails corresponding to the motion of species  $A$  overlap the blue trails representing the motion of species  $B$ , but the region originally occupied by species  $A$  contains no blue trails.

One issue is whether the results reported here apply more generally for other types of particle interactions. We considered only Yukawa interactions; however, the meandering channel structures which lead to the mixing are a universal feature of one-component systems undergoing plastic flow through random quenched disorder. Studies performed on systems with long-range logarithmic interactions [17] as well as short range interactions [24] which show this plasticity lead us to believe that plastic flow generated by random disorder can produce enhanced mixing for a wide range of particle interactions. For our specific system of Yukawa particles, experiments on single component systems have already identified a channel-like plastic flow regime [9].

In summary, we have shown that two-dimensional plastic flow induced by quenched disorder in the absence of thermal fluctuations can lead to efficient mixing and enhanced diffusion in phase separating systems. This mixing occurs due to the meandering of particles through

riverlike flow structures. We map the general mixing phase diagram and find that mixing is optimized in regimes where the particles depin in an intermittent fashion. For higher external drives the mixing is strongly reduced. These results should be general to a variety of systems where meandering flow channels appear.

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